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Hardening of the Coulomb gap by electronic polarons

R. Chicon and M. Ortuño
Departamento de Física, Universidad de Murcia, 30001 Murcia, Spain

M. Pollak
Department of Physics, University of California–Riverside, Riverside, California 92521

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A one-electron density of states for a system of interacting electrons is obtained in the limit of strong localization. The method is based on making the ground state stable with respect to electronic polaron excitations. The resulting density of states presents a relatively small hard gap of the order of a fifth of the Coulomb gap.

I. INTRODUCTION

The ground-state properties of highly disordered systems can be strongly affected by the long-range Coulomb interactions. The importance of the interactions increases as the system approaches the metal-insulator transition from the metallic side, and is particularly strong on the insulating side of the transition. There the Coulomb interactions drastically change the electronic structure and properties of the system.

One of the most important effects is the depletion of the single-particle density of states (DOS) near the chemical potential,[1] referred to as the Coulomb gap.[2] Efros and Shklovskii[2] obtained a universal parabolic DOS around the chemical potential by making the ground state stable against one-electron transitions. Efros,[2] Baranovskii *et al.*,[4] and Davies[5] studied the consequences of stabilizing the ground state against certain many-electron transitions, obtaining hard gaps (i.e., gaps stronger than parabolic) in the DOS. These authors consider transitions in which an electron is brought from (or taken to) infinity to (from) a site i near the chemical potential, where it can interact with other electrons. Of particular importance are nearby electrons capable of low-energy excitation. These low-energy excitations are transitions to nearby vacant sites. Such compact electron-hole pairs are not depleted by the Coulomb gap. Their effect on the electron inserted in (deleted from) site i can be represented approximately as a monopole-dipole interaction.

In this paper we want to explore the consequences of stabilizing the ground state against more general, many-electron transitions. Specifically, we consider the transitions called in the literature[3] polaron excitations. These are transitions where one electron makes a long but finite hop, while others make short hops near the initial and final sites of the long hop.

It should be mentioned that many-electron excitations of another type were argued to be more important at low excitation energies.[6] These so-called cascade excitations are characterized by transitions of a number of electrons, all of comparable length, and with comparable spacings between the individual transitions. Computer simulations confirmed the importance of such low-energy cascade excitations.[7] However, there may not be an overall consensus on their importance, and the statistics of the cascade transitions is also not adequately known. In this paper we therefore focus on polaron excitations, and calculate how they affect the one-particle density of states in the Coulomb gap.

Although the treatment here is of general applicability to strongly localized systems, we will concentrate for the sake of concreteness on systems exemplified by lightly doped crystalline semiconductors where double occupation of sites is forbidden, and where quantum effects can be neglected, since the tunneling from site to site is extremely small. The Hamiltonian[2] then includes only contributions from the Coulomb interactions and from the random site energies. The latter are assumed to be uniformly distributed over a range W, and uncorrelated from site to site. The single-particle energy E_i associated with site i is defined as the sum of the random energy of site i plus the Coulomb interaction energy with the electrons in all other sites. The one-particle DOS is defined as the density of these energies per unit energy when the occupation is that of the ground state.

The paper is organized as follows. In Sec. II we calculate an upper bound for the energy distribution of electronic polarons as a function of the length of the long jump. In Sec. III we apply the previous results to calculate the density of long low-energy one-electron excitations. The electron-hole correlation function is obtained in Sec. IV and the density of states near the chemical potential in Sec. V. Some conclusions are presented in the last section.

II. ENERGY DISTRIBUTION OF ELECTRONIC POLARONS

In this section we first calculate the probability distribution for the energy relaxed by the system in response to a single long electronic hop of length R. As this energy cannot be greater in magnitude than the excitation energy of the long hop for the ground state to be stable, this results in an estimate of the density of excitations as a function of energy, at a fixed R. This is the natural extension of previous calculations[5] for infinite hops, in which the distribution for the energy relaxed by the system results in an estimate of the density of states. This estimate of the DOS is considered in the literature as an upper bound for the DOS. The rationale is that not all possible transitions that stabilize the ground state (e.g., cascade transitions) are taken into account. However, we believe that correlations between relaxing pairs, which are neglected in this treatment, can produce the opposite effect. Thus, the consideration of these estimates of the DOS as upper bounds should be taken with care.

As will be seen below, the constraints for finite R mentioned above introduce additional correlations in the electron-hole separation from those present in the model of Efros and Shklovskii. The correlations produce a depletion around any given site of states with opposite occupancy. The effective excluded volume, defined as the average size of this depletion region, around a site close to the chemical potential becomes greater, which results in a smaller DOS than the parabolic DOS predicted by Efros and Shklovskii.[2]

When a hop of length R occurs, the pairs whose excitation energy is smaller than their interaction energy with the electric field produced by the transition will polarize. This latter field is the difference between the fields of the final and initial configurations of the charges, which for a finite one-electron jump is the field produced by a positive charge at the original site and a negative charge at the final site.

Let us call $-x$ the energy relaxed by the system, i.e., by the set of short pairs that polarize. The distribution of the values of x will be calculated later. The total energy involved in the process is then (we use units in which $e=1$)

$$\Delta E = E_j - E_i - 1/R - x \ , \tag{1}$$

where E_j and E_i are the energies of the initial and final sites of the long hop. The sign of x has been chosen such that $x>0$. For the ground state to be stable, ΔE must be greater than zero, so that only those long hops with a relaxation energy smaller than the energy of the hop, $E_j - E_i - 1/R$, are allowed.

Since we deal with a disordered system, x is a random variable, with a probability density $g(x \mid R)$. Long hops between regions where the density of low-energy pairs is small due to fluctuations cannot relax much energy and will therefore contribute to $g(x \mid R)$ at small x. In accordance with what was said earlier, we will consider $g(x \mid R)$ as an estimate of the density of excitations, as a function of energy, at a fixed R. We shall first calculate $g(x \mid R)$. The consequences of the interpretation of $g(x \mid R)$ will be analyzed in the next section.

We assume here that the low-energy pairs are uniformly and isotropically distributed in space, with n pairs per unit volume. The length r and energy w of the pairs are distributed according to a probability density $f(w,r)$. The interaction of the pairs with the electric field is approximated as an interaction of point dipoles. This is justified because the relevant low-energy pairs are very short. The most restrictive assumption we make here is that the interaction between pairs is neglected as far as the relaxation process is concerned. The assumption seems reasonable because of the shorter range of the dipole-dipole interaction, compared with the Coulomb interaction between charges. The assumption allows us to calculate the distribution of the energy, relaxed by the system in response to the hop of length R, from the distribution of relaxation energies due to one dipole. The function $g(x \mid R)$ is the convolution of M functions $\tilde{g}(x \mid R)$, which denotes the distribution of the relaxation energy by one dipole, and M is the total number of dipoles. It is convenient to perform a Laplace transform, since the transform of the convolution is a product of transforms. This procedure follows the method of Chandrasekhar,[8] also applied to this problem by Davies.[5]

Calculating $\tilde{g}(x \mid R)$ for the case of a long hop of length R gives

$$\tilde{g}(x \mid R) = V^{-1} \int \int \int d^3l \, d^3r \, dw \, f(w,r) \times \delta(x + w - \mathbf{r} \cdot \mathbf{E}(\boldsymbol{l})) \ . \tag{2}$$

Here V is the volume of the system, and $\mathbf{E}(\boldsymbol{l})$ is the electric field associated with the finite hop, i.e., the field of a positive charge and a negative charge separated by a distance R. For the distribution of low-energy excitations, $f(w,r)$, we take a separable form, $f(w,r)=f(r)/W_1$, with w varying from 0 to W_1. This is justified because the range for the excitation energy of the dipoles of interest is very small. In this we follow Davies,[5] but use a more refined $f(r)$ which takes into account electron-hole correlations, as will be explained later.

The calculation of $\tilde{g}(x \mid R)$, as well as of its Laplace transform, can be performed analytically, using the separable form for $f(w,r)$. However, the inverse Laplace transform, needed to obtain $g(x \mid R)$, cannot be performed analytically. A practical approximation for calculating $g(x \mid R)$ which we use is a separation into two limiting cases: $g_D(x \mid R)$ for small x, where the field of the hop is approximated by the field of a point dipole of magnitude R, located at the center of R, and $g_M(x \mid R)$ for large x, where the field is represented by a field of two independent monopoles of opposite sign. The rationale is that low relaxation energies correspond to large spatial regions (of order R) depleted of low-energy pair excitations. The pairs outside the region then see the two charges roughly as a point dipole. On the other hand, a large energy is relaxed when low-energy excitations are near the sites involved in the long jump, so they will see a monopolar field. The value x_C of x which separates between $g_M(x \mid R)$ and $g_D(x \mid R)$ is taken to be $g_M(x_C \mid R)=g_D(x_C \mid R)$, i.e.,

$$g(x \mid R) = \begin{cases} g_D(x \mid R), & x < x_C \ , \\ g_M(x \mid R), & x > x_C \ . \end{cases} \tag{3}$$

The function $g_M(x \mid R)$ can be directly obtained from the distribution for the energy relaxed by a single monopole, calculated by Davies,[5] and is given by

$$g_M(x \mid R) = A \left[\frac{x}{2E_D} \right]^{-3/2} \exp\left[-\frac{2E_D}{x} \right] , \tag{4}$$

where

$$A=\frac{\exp\{2[E_D/(\pi W_1)]^{1/2}\}}{\pi^{1/2}W_1} \tag{5}$$

and

$$E_D=\left[\frac{\pi}{4}\frac{8\pi}{15}\frac{n}{W_1}\langle r^{3/2}\rangle\right]^2 . \tag{6}$$

n/W_1 represents the density of very-low-energy excitations which contribute to this relaxation mechanism. $\langle r^{3/2}\rangle$ is the mean value of $r^{3/2}$, averaged over the pairs considered.

The calculation of $\tilde{g}_D(x\mid R)$ can be performed analytically, obtaining

$$\tilde{g}_D(x\mid R)=V^{-1}\frac{\pi A'R}{6W_1}\langle r\rangle\left[\frac{1}{x}-\frac{1}{x+w}\right] , \tag{7}$$

where $\langle r\rangle$ is the mean value of r over the considered pairs and $A'=2+(\frac{1}{3})^{1/2}\ln(2+3^{1/2})$. As mentioned before, the Laplace transform $G_D(s\mid R)$ of the function $g_D(x\mid R)$ is the product of M Laplace transforms of $\tilde{g}_D(x\mid R)$. In our case, this can be written in the form

$$G_D(s\mid R)=\left[1-V^{-1}\int_0^\infty dx[1-\exp(-xs)]V \times\tilde{g}_D(x\mid R)\right]^M . \tag{8}$$

Substituting Eq. (7) in Eq. (8) and taking the limit $M,V\to\infty$ so that $M/V=n$, we finally get

$$\ln(G_D(s\mid R))=-\frac{n}{W_1}\frac{\pi A'R}{6}\langle r\rangle \times[\gamma+\ln(W_1 s)+\exp(W_1 s)E_1(W_1 s)], \tag{9}$$

where γ is Euler's constant and $E_1(z)$ is the exponential integral defined in Abramowitz and Stegun.[9]

The inverse transform of $G_D(s\mid R)$ can be performed easily for small values of x, which is the regime of interest for us and corresponds to large values of s. In this limit we have

$$g_D(x\mid R)=\frac{e^{-B\gamma}}{W_1^B}\frac{x^{B-1}}{(B-1)!} , \tag{10}$$

where

$$B=\frac{\pi A'\langle r\rangle R}{6}\frac{n}{W_1} . \tag{11}$$

Since we allow for the simultaneous polarization of many low-energy excitations, we have to consider an appropriately disjointed set of pairs[10] in place of all the possible short pairs which could be formed from N sites. By disjointed set we mean here a partitioning into pairs where each electron is assigned a hole, and vice versa. There are many possible ways to partition the pairs. Here the partitioning is done in accordance with the excitation energy. The many-electron transition of interest is that which has the smallest total excitation energy (the "best" transition). To approximate this transition we partition the pairs in such a way that each electron is associated with the hole which gives the smallest one-electron excitation energy ("best" pair). Because of frustration such pairs are not generally disjointed, since two electrons may use the same hole. For disjointed pairs we must then use also "second-best" pairs, where an electron uses the hole which gives the second lowest one-particle excitation. The average value $\langle r\rangle$ and the density of pairs per unit energy at low energy n have to be calculated from the disjointed set of pairs.

The essential features of the calculation of the density of disjointed pairs are as follows. First, we obtained a joint probability density of electron-hole excitations $n(E_0,E,r)$ as a function of the energy of the occupied site E_0, the energy of the empty site E, and their separation r. To obtain it we used an electron-hole pair correlation function $n(r)$, which represents the fraction of unoccupied sites at a distance r to a given occupied site, obtained from computer simulation, and took into account the stability of the ground state and the properties of short pairs. Using Poisson statistics we calculate from $n(E_0,E,r)$ the distribution of best and second-best pairs. As mentioned above, only a fraction of the best pairs belongs to the best set of disjointed pairs, and the same is true for second-best pairs. For our distributions the fractions are $\frac{1}{2}$ and $\frac{1}{4}$, respectively.[10]

In Fig. 1 we show the results for the distribution of energy relaxed, $g(x\mid R)$. The dashed line corresponds to the monopolar approximation, i.e., when the relaxation is produced by two independent monopoles. The solid lines correspond to $g_D(x\mid R)$ for different values of R. For a given R we set $g(x\mid R)=g_D(x\mid R)$ for $x<x_C$, and $g(x\mid R)=g_M(x\mid R)$ for $x>x_C$, where $g_M(x_C\mid R)=g_D(x_C\mid R)$. We have chosen the unit of length equal to the intersite separation a, and the unit of energy equal to the Coulomb interaction between sites separated by a distance a. We have considered a disorder energy range W equal to unity in these units.

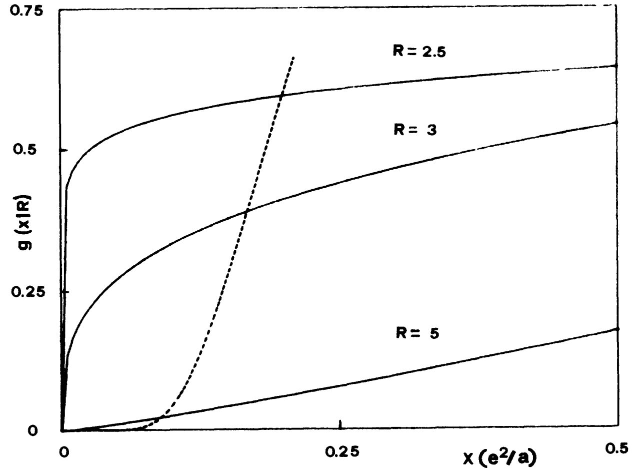

FIG. 1. Probability density $g(x\mid R)$ for the energy relaxed by the system in response to a finite hop of length R: dipolar approximation (solid lines), for several values of R, and monopolar approximation (dashed line).

III. DENSITY OF LONG ONE-ELECTRON EXCITATIONS

Since the energy of any transition from the ground state has to be positive, ΔE in Eq. (1) has to be greater than zero, and therefore $g(x \mid R)$ represents an estimate of the density of one-particle excitations for a given R, in the same way that the distribution of the energy relaxed by an infinite jump provided an estimate of the density of states. To be more precise, we can interpret $g(x \mid R)$ as an estimate of the probability density that, given an empty and an occupied site separated by a distance R, the sum of their site energies (measured in absolute value from the chemical potential) is $x+1/R$. With this interpretation, we can notice from Fig. 1 that, for small R, $g(x \mid R)$ introduces no restriction on the density of pairs, in agreement with the results for the DOS considering a dipole-dipole interaction.[4] This criterion must be ultimately verified by the true density of excitations. An approximate density of excitations, $n_2(w,r)$ (w being the excitation energy of the pair, not the sum of the site energies, and r the length of the pair), can be obtained from the probability density $n(E_0,E,r)$, as will be briefly described below. This density will not incorporate the effects of relaxation, which will be introduced explicitly later. For large enough r, $n_2(w,r)$ is given by

$$n_2(w,r)=N^2 n(r)\tfrac{32}{30}(w+1/r)^5\,, \qquad r>1/W,\quad w<\Delta-1/r\,, \tag{12}$$

where $\Delta=1/(2W^{1/2})$ is the width of the Coulomb gap and N is the density of sites. This expression for $n_2(w,r)$ corresponds to the self-convolution of the parabolic DOS, taking into account the excitation term. This expression is not adequate for the problem of the calculation of the energy relaxed by the system, because in that problem short pairs are the relevant ones.

Thus, the more refined expression that we take for the density of excitations in the region of large separation and low excitation energy is given by

$$\tilde{n}_2(w,r)=\frac{N^2}{2}n(r)\min[\tfrac{32}{15}(w+1/r)^5, g(w \mid r)]\,. \tag{13}$$

From the form of $g(x \mid R)$ we can notice that the probability of finding an electron-hole pair with a large separation and a low excitation energy is very small. This means that it is very unlikely for an electron and a hole with given site energies to be separated by a distance slightly above the minimum distance obtained in the parabolic model for the DOS.

IV. ELECTRON-HOLE CORRELATION FUNCTIONS

To calculate the increase in the effective excluded volume around a site due to relaxation, useful in the calculation of the DOS, it is convenient to analyze the correlation function $f_\varepsilon(r)$. This is defined as the actual density of electron-hole pairs, for a given $\varepsilon=E_i+E_j$, as a function of the distance r, divided by the average density of such pairs. In the model of Efros and Shklovskii the correlation function has the simple form

$$f_\varepsilon(r)=\begin{cases}0, & r<r_{\min}\,,\\ 1, & r>r_{\min}\,,\end{cases} \tag{14}$$

where $r_{\min}=1/\varepsilon$. Considering the effects of relaxation associated with a long hop results in a decrease of the correlation function for distances just above $r_{\min}$. In our model we have

$$f_\varepsilon(r)=\begin{cases}0, & r<r_{\min}\,,\\ \dfrac{\tilde{n}_2(\varepsilon-1/r,r)}{n_2(\varepsilon-1/r,r)}, & r>r_{\min}\,.\end{cases} \tag{15}$$

In Fig. 2 we show the correlation function $f_\varepsilon(r)$ for three values of ε: 0.1, 0.16, and 0.2 in our units. The dashed line represents the results in the model of Efros and Shklovskii.[2] Our result for $f_\varepsilon(r)$ corresponds to the

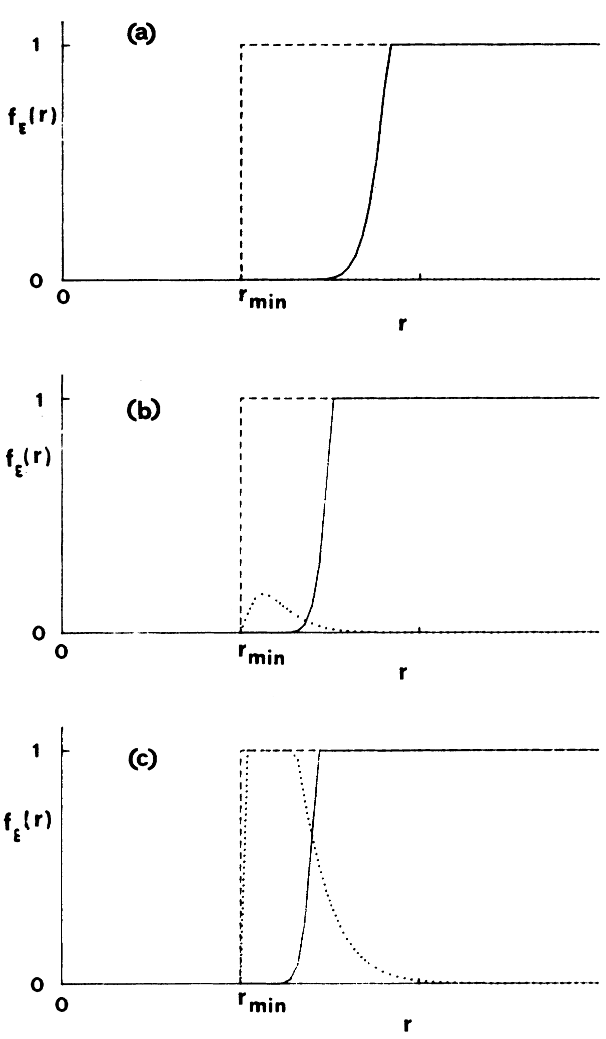

FIG. 2. Correlation function $f_\varepsilon(r)$ for three different values of ε: (a) 0.1, (b) 0.16, and (c) 0.2. For each r, $f_\varepsilon(r)$ is the maximum between the dotted line (dipolar approximation) and the solid line (monopolar approximation), saturating at unity. The dashed line corresponds to the results in the model of Efros and Shklovskii.

envelope of the dotted line [contribution from the dipolar approximation to $g(x \mid R)$] and of the solid line (monopolar approximation), and by the condition $f_\varepsilon < 1$. At very low energies [Fig. 2(a)] only the monopolar contribution to $f_\varepsilon(r)$ is relevant, since the field is essentially that of two monopoles and the dipolar part is negligible at all distances. In Fig. 2(b) we see how the dipolar contribution plays an important role for distances close to $r_{\min}$. This is true for ε higher than 0.16, for the range W of disorder energies considered. At large ε [Fig. 2(c)] we notice how the dipolar contribution suppresses the decrease in $f_\varepsilon(r)$ arising from the monopolar part, such that the correlation function of Efros and Shklovskii[2] is recovered. The energy range over which the dipolar contribution is small enough to suppress $f_\varepsilon(r)$ significantly below Efros and Shklovskii is quite small, of the order of 0.2 in our units.

The nonmonotonic behavior of $f_\varepsilon(r)$ is an artifact of splitting $g(x \mid R)$ into strictly dipolar and monopolar expressions. A smoother interpolation would eliminate this slight anomaly. In any case, this is of very little practical importance in the calculation of the DOS, due to the very small energy range over which the dipolar contribution is neither negligible nor dominant.

It should be noted that the abscissa scale is different in each part of Fig. 2, since $r_{\min}$ is a function of ε.

V. HARD GAP IN THE DENSITY OF STATES

From the correlation function $f_\varepsilon(r)$ it is possible to obtain an excluded volume around a site, and from it to calculate the DOS. The excluded volume around a site with energy E is given by

$$V_{\rm ex}(E)=\int_{R^3} d^3r[1-f_\varepsilon(r)] \ . \tag{16}$$

Strictly speaking, this excluded volume is with respect to sites with opposite occupancy to that of the original site, and with energy $\varepsilon - E$. One could produce a self-consistent method to calculate the DOS from this exact interpretation of $V_{\rm ex}(E)$.[3] The results of this refined method are expected to be the same, except for a proportionality constant, as those obtained by simply considering $\varepsilon = 2E$ and interpreting $V_{\rm ex}(E)$ as an effective-energy-dependent excluded volume.[2]

Once the excluded volume has been obtained, the calculation of the DOS is performed through the expression

$$N(E)=\frac{1}{6}\frac{d}{dE}\left[\frac{1}{V_{\rm ex}(E)}\right] . \tag{17}$$

The factor $\frac{1}{6}$, obtained by Efros,[3] takes into account the self-consistency of the problem.

Before showing the results for the DOS obtained from Eq. (17), we would like to consider a simple model which contains some of the interesting physics of the problem. Let us assume that the relaxation by the short pairs is at least x_0. Then $g(x \mid R)$ would be zero for $x < x_0$ and very large for $x > x_0$ (this corresponds to the extreme case in which we only have a monopolar contribution to $g(x \mid R)$, and this presents a "completely hard" gap). In this case, for $\varepsilon < x_0$ we have $f_\varepsilon(r)=0$ for all distances, and thus the DOS is strictly zero for energies smaller than $x_0/2$. For $\varepsilon > x_0$, the correlation function $f_\varepsilon(r)$ is a step function, as in the model of Efros and Shklovskii, but with a modified $r_{\min}$, given now by $r_{\min}=1/(\varepsilon - x_0)$. This would result in a displaced parabolic DOS:

$$N(E) \propto (E-x_0/2)^2, \quad E > x_0/2 \ . \tag{18}$$

This simple model represents quite well our quantitative results in the range of very low energy, where typical distances are large and the dipolar contribution to $g(x \mid R)$ is negligible compared to the monopolar one.

In Fig. 3 we show the results for the DOS (relative to the unperturbed DOS g_0, i.e., that without interactions) obtained using Eqs. (15) and (16) in Eq. (17) (solid line). For comparison, we also show the parabolic DOS (dashed line). The calculated DOS presents a small hard gap, of the order of a fifth of the Coulomb gap, compatible with the results of computer simulations.[11] The DOS is strictly zero in a small, but finite, interval around the Fermi level, corresponding to the range in which there is no dipolar contribution to $g(x \mid R)$. For higher energies the dipolar contribution becomes rapidly dominant and we recover the original parabolic DOS. The small energy range over which the DOS goes from the hard gap to the parabolic DOS has its origin in the rapidity with which the dipolar contribution to $g(x \mid R)$ takes over, after it appears.

VI. CONCLUSIONS AND SUMMARY

In the preceding sections we studied the consequences of the stabilization of the ground state of interacting electrons in disordered insulators against polaron excitations. The main approximations inherent in this treatment were the following. It was assumed that the important mechanism for the hardening of the Coulomb gap is by polarization of compact low-energy one-electron excitations, as has also been done in previous works.[3,4,5] The approximate nature of this assumption lies in the fact that relaxa-

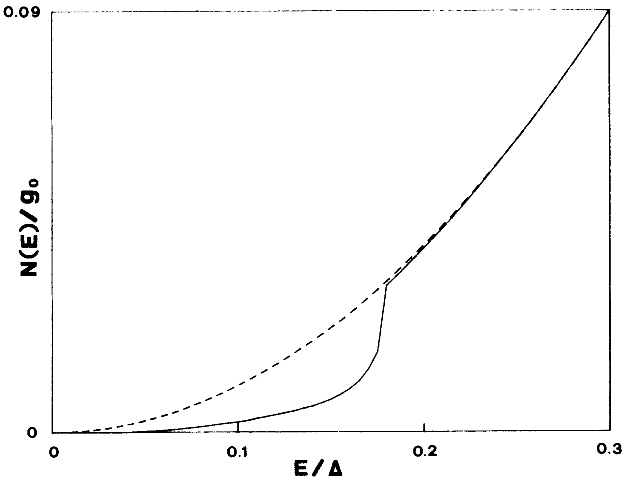

FIG. 3. DOS, relative to the unperturbed DOS g_0, obtained considering the relaxation of the system in response to finite hops (solid line), and parabolic DOS (dashed line).

tion by low-energy many-electron excitations may be more important, in part because of their huge number. Preliminary results on the simultaneous low-energy two- and three-electron excitations seem to indicate that their importance is in fact comparable to that of low-energy one-electron excitations.[12] The effect of the n-electron excitations could be partially simulated by an effective new density of one-electron excitations, if their importance is limited to a relatively small n, and if their statistics becomes better known.

Considering that the function $N(E)$ given by Eq. (17) is an upper bound for the DOS implicitly assumes that there is no clustering. That is to say that sites with the same occupation and with energy close to the chemical potential are essentially independent and do not correlate to form clusters. Also, the distribution of relaxing pairs considered is not independent of the stabilization of the ground state against other transitions, which could prevent our estimate from being an upper bound.

The contribution of one-electron transitions to the ac conductivity at $T=0$, assuming a parabolic DOS, turns out to be linear in frequency, with a logarithmic correction at low frequencies. This results in a divergence in the static dielectric constant, as can be seen from the Kramers-Kronig relations. A gap harder than parabolic, such as the one we have obtained, removes such a divergence. Furthermore, the electron-hole correlation function obtained in Sec. IV makes the calculation of the conductivity and dielectric constant more realistic.

In summary, in this paper the single-particle DOS for interacting localized electrons is obtained by stabilizing the ground state to polaron excitations, where an electron makes a long jump, and the rest of the system relaxes. The distribution of relaxation energies results in an estimate of the density of long low-energy one-electron excitations. The stability condition against the more general excitations considered here produces an increase in the separation between electrons and holes near the chemical potential, with respect to the separation in the model of Efros and Shklovskii. Through the evaluation of the electron-hole correlation function we calculate the effective excluded volume around each site, and from this an estimate of the DOS. We find that stability of the ground state with respect to polaron excitations results in a hard gap, of the order of a fifth of the Coulomb gap.

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